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## **EVALUATION OF IMPROVED TRANSPARENT MATERIALS AND ADHESIVES FOR BALLISTIC AND IMPACT SHIELDS**

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**AD No.**

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EVALUATION OF IMPROVED TRANSPARENT MATERIALS  
AND ADHESIVES FOR BALLISTIC AND IMPACT SHIELDS

G. L. Ball III  
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I. O. Salyer

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## FOREWORD

This project was carried out in the Polymer Applications Section of the Dayton Laboratory of Monsanto Research Corporation, Dayton, Ohio, under Air Force Contract F33615-67-C-1315.

This contract was initiated under Project Number 7381, "Materials Application," Task 738108, "Application of Materials and Processes for Tactical Warfare." This program is administered by the Air Force Materials Laboratory, Air Force Systems Command, Wright-Patterson Air Force Base, with Mr. R. E. Wittman as Project Engineer.

This work was conducted under the technical direction of Mr. I. O. Salyer, with Mr. G. L. Ball III functioning as Project Leader. The Principal Investigator on the project was Mr. A. Wojtowicz, who was assisted by Mr. R. D. Myers and Mr. T. J. Bucher.

This report describes work performed from 1 June 1967 to 31 December 1968 and was submitted for approval in November 1969.

Acknowledgement is given to Monsanto Company and to Libbey-Owens Ford Company for their support in supplying some necessary materials at no cost to the program.

This technical report has been reviewed and is approved.

*Albert Olevitch*  
Albert Olevitch, Chief  
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## ABSTRACT

A program was conducted to make available to the Air Force the most advanced materials for use in transparent armor. The materials of interest were a high strength aluminum borosilicate glass, a high clarity polycarbonate, and transparent adhesives for bonding the glass to the polycarbonate.

The aluminum borosilicate glass was made available in various thicknesses up to 1/2 inch. The polycarbonate was an injection moldable grade described as Merlon<sup>®</sup> E-271. It was necessary to determine injection molding cycles and to injection mold 12 inch by 12 inch plates in thicknesses from 1/8 inch up to 1/2 inch.

Two transparent adhesives were examined. These consisted of a modified polyvinylbutyral (PVB) from Monsanto Company and a thermoplastic urethane from Mobay Chemical Company. Unfortunately, neither adhesive was found to be suitable for the application since haze was evident in laminates and could not be removed. Efforts to refine either one of these materials were discontinued since a more promising material became available at the end of the program.

A minor amount of ballistic testing was conducted on the polycarbonate and significantly it was shown that ballistic performance was not related to low rate impact properties. The implication of these results is that orientation of a thermoplastic is of little benefit and annealing is not detrimental to ballistic performance. It was also shown that a single homogeneous plate had better ballistic performance than multiple plies of polycarbonate.

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SECTION I  
INTRODUCTION

Armor is a significant item in maintaining the usefulness of military aircraft. Transparent armor is especially important. Owing to the critical needs in southeast Asia and a most unique combination of effort by the U.S. Army, U.S. Air Force, U.S. Navy and U.S. Marine Corps in providing effective armor on a joint basis, the work of the Department of Defense, DOD contractors, defense suppliers, and industry have made available some new and improved materials.

The program conducted under this contract was designed to make these latest advances in materials known to the Air Force, to demonstrate the usefulness of this technology, and to evaluate the systems so that the latest art would be available for incorporation into Air Force systems. Monsanto Research Corporation therefore procured, designed, and/or fabricated ballistic and Mach I impact shields incorporating techniques and materials developed by other DOD contractors, commercial organizations, and our own laboratories.

## SECTION II

### SUMMARY AND CONCLUSIONS

The experimental program was designed to provide the best available impact-resistant transparent armor materials to the Air Force, based on their criteria for selection. The Air Force is primarily interested in achieving the best possible level of defeat with a given weight of material. Transparency and freedom from distortion are also of considerable added importance because the transparent materials are intended for use in aircraft windshields.

The program that was conducted was based on the availability of a new, modified polyvinylbutyral (PVB) adhesive that reportedly would bond to and be compatible with polycarbonate and glass. This was considered significant since commercial-grade PVB crazes polycarbonate surfaces by extraction of a plasticizer incorporated into the PVB. It was anticipated that high-impact polycarbonate could be used in place of stretched acrylic if this modified PVB performed as expected. The more optically transparent polycarbonates that had become available were also of interest for incorporation into this system.

Accordingly, laminates consisting of high clarity polycarbonate and high strength aluminum borosilicate glass were prepared using the modified PVB adhesive. However, when these laminates were prepared, the desired improvement was not demonstrated. At best, polycarbonate laminates bonded with the modified PVB exhibited a moderate haze. The crazing that occurred was much less than that normally encountered with commercial PVB, and thus a significant improvement was demonstrated. The system, however, could not be considered adequate for aircraft windshield applications.

A thermoplastic urethane was also investigated as an adhesive for bonding polycarbonate to glass. The haze that was evident in laminates prepared using the commercially-available compositions, however, was more than could be tolerated. Therefore, this approach was also abandoned.

At the end of the program, a series of ethylene terpolymers became available. On preliminary analysis, these looked most promising as adhesives. Although no work was done with these ethylene terpolymers, they are being evaluated for clarity, bonding, and compatibility with polycarbonate under a subsequent contract.

A major portion of the program involved the molding and characterization of the more recently available high-clarity polycarbonate. This work was required because high-clarity polycarbonate (Mobay Chemical Company E-271) was not available in extrudable form, but required injection molding. An injection molding program, therefore, involving the design and use of a 12 in. X 12 in. plate injection molding die, was conducted. The die, constructed by Monsanto Research Corporation (MRC), was designed to incorporate inserts for molding parts in thicknesses up to 1/2 inch.

Molding conditions were established for preparing Merlon E-271 polycarbonate plates from 1/8 in. to 1/2 in. thick. This required the development of a different set of molding conditions for each thickness. Since the polycarbonate plates were prepared for evaluation with respect to optical transparency, it was necessary that they be molded relatively strain free and with undistorted surfaces. Development of injection molding cycles that incorporated hot die molding techniques were thus needed. The hot die molding technique consisted of injecting the polycarbonate into the mold above its glass transition temperature ( $300^{\circ}\text{F}$ ) followed by cooling and removal of the part. Such a technique required molding cycle times of 15 to 30 min.

The optical transparency and the low rate impact properties of the molded polycarbonate were determined. The optical transparency measurements showed that the E-271 polycarbonate exhibited from 5 to 15% (on an absolute basis) increase in transparency over commercial grades. The impact properties of the polycarbonate were in the range anticipated for a polycarbonate, i.e., 15 ft-lb/in. of notch.

The laminating of polycarbonate to glass with various adhesives required autoclaving through a thermal cycle that included temperatures close to the transition region of polycarbonate. Since these temperatures could anneal and remove the characteristic high impact strength of polycarbonate, the impact properties as a function of annealing temperature were determined. It was found that annealing the polycarbonate at temperatures up to  $200^{\circ}\text{F}$  had no effect, while annealing at temperatures above  $200^{\circ}\text{F}$  caused reduction in the impact strength of polycarbonate to a level of 2 ft-lb/in. of notch. A simple Vicat indentation test was shown to be valuable in determining the degree of annealing in the polycarbonate.

The optical properties of polycarbonate-glass-laminates were determined to test the compatibility of the high-clarity polycarbonate and proposed bonding adhesives. As noted above,

the prime candidates for use as adhesives were somewhat disappointing, but the transparency of the glass and polycarbonates was determined.

The V50 ballistic limit and merit ratings were obtained for the injection-molded polycarbonate using caliber .30 ball M2 and AP projectiles. Two major conclusions were reached. First, the ballistic performance of the polycarbonate increased as the homogeneity of the composite increased; i.e., in a given total thickness, the fewer the number of plates, the better the ballistic performance. The second and more significant conclusion was that the ballistic performance of annealed polycarbonate, in which the Izod impact properties were significantly reduced, was no different from that of unannealed polycarbonate. This finding was most important and confirmed some work conducted at about the same time by the U.S. Army Natick Laboratories. Their work showed that stretched or biaxially oriented polycarbonate offered no significant advantage over unoriented polycarbonate in ballistic performance.

Data showing the relationship between the thickness of polycarbonate (areal density) and its V50 ballistic limit were determined. Since no laminates which exhibited sufficient optical transparency were prepared, none were tested.

The major conclusions obtained from this work are:

- A new ethylene terpolymer adhesive for bonding polycarbonate is potentially available,
- A modified polyvinylbutyral and a commercial thermoplastic urethane are not suitable for bonding polycarbonate,
- A high-clarity polycarbonate is indeed available,
- High-clarity polycarbonate plates can be injection molded using a sophisticated hot-die technique,
- The ballistic performance of polycarbonate is not related to its low-rate impact properties, and
- The ballistic performance of polycarbonate is related to its homogeneity.

## SECTION III

### EXPERIMENTAL PROGRAM

#### 1. GENERAL

The objective of the experimental program was to procure, design, and/or fabricate transparent ballistic and aircraft impact shields. The shield materials were to incorporate the best available high-strength glass, a newly available high-clarity polycarbonate, and transparent adhesives to bond the polycarbonate to the glass (which of necessity must be compatible with the polycarbonate). While polycarbonate has excellent and known high impact characteristics, high clarity (especially as compared to stretched acrylic) has been lacking, and good adhesives that do not attack the polycarbonate have not been readily available.

Our experimental program to examine this system for the Air Force and to make it available for use in aircraft involved:

- A limited literature survey on transparent materials,
- Molding of high-clarity polycarbonate plates,
- Characterization of the high-clarity polycarbonates,
- Laminating of transparent shields, and
- Ballistic characterization and analysis.

The major portion of this program involved the injection molding of high-clarity polycarbonate plates and physical and ballistic characterization of them. This molding was necessary because the high-clarity polycarbonate was not readily extrudable as are commercially available polycarbonates. Lamination of these polycarbonate plates to glass and ballistic characterization to determine adhesive qualities were then conducted.

#### 2. LITERATURE SURVEY

A literature survey was conducted, and abstracts and pertinent literature on transparent armor systems were compiled. The fact that some of the more promising information had become available only more recently and for the most part was classified "Confidential" somewhat hindered the ready exchange of information. However, a limited survey was achieved.

The available literature was listed according to author, title, date, AD number, source of work, sponsoring agency, and contract number. This information was punched into IBM cards to permit additional entries and search.

Even though some of these reports were unclassified, the combination of the various titles into one list required that the total document be classified. This list was distributed to the Air Force as part of a Confidential monthly report. Since the other experimental work conducted in this program was not classified, we have chosen to not include the literature survey in this report. The information is available for further reproduction and distribution as required and is also available for use on Contract Number F33615-69-C-1190.

### 3. INJECTION MOLDING OF HIGH-CLARITY POLYCARBONATE PLATES

Injection molding of the high-clarity polycarbonate plates in various thicknesses required design and modification of a 12 in. X 12 in. die with inserts to permit the preparation of plates, the establishment of injection molding conditions for molding the plates flat with an optical surface, and the molding of a quantity of plates of various thicknesses.

#### a. Design and Modification of the Plate Injection Molding Die

To provide 12 in. X 12 in. polycarbonate plates of various thicknesses it was necessary to consider a mold design including exchangeable inserts. Monsanto Research Corporation made available to the program a die base as well as incorporated gates and runners necessary to provide proper functioning of the die. The die was designed to be installed in an HPM 20-ounce injection molding machine that was made available to the program by the Atomic Energy Commission.

A most unique factor about this injection molding die was the consideration that it provide for the molding of parts of various thicknesses from 0.125 in. up to 0.5 in. Most dies are designed to produce one particular part whose dimensions are seldom changed, especially in terms of the amount of polymer required to fill the mold, and therefore flow considerations are fixed. For the molding of various-sized parts, some versatility in the flow regime must be available so that machine parameters can be adjusted for these different degrees of fill.

Our experimental program called for the examination of 10 in. X 10 in. plates of molded materials that were to be relatively free of surface imperfections. Since knockout pins are required to remove parts from a mold, we designed a 12 in. X 12 in. plate from which the 10 in. X 10 in. plate could be cut. The mold faces were polished to impart a smooth surface to the molded polycarbonate plate.

The runner and gating system was designed to provide either center or edge gating of the polycarbonate plate. To achieve the minimum degree of strain and to reduce the time necessary to establish molding conditions for the parts, the center gate was used. This type of gating resulted in an imperfection in the center of the part (a hole where the gate was drilled out). If desired, this gate could be removed and polished to maintain the continuity of the part. When the surface is laminated to another part, this imperfection is invisible. The die could also be prepared (but was not so used) to allow for corner or edge gating, so that both surfaces would be entirely free from imperfections.

To provide for the variable thicknesses and for alternative means of filling the part, the die was designed as a 3-plate mold. The third plate consisted of a floating plate into which the runners for either center or edge gating were included. These runners and gates could be used in combination or individually, or could be entirely eliminated by blank inserts. This configuration is shown in Figure 1.

Because of the anticipated difficulty of molding such a large part in a 20-ounce machine, the center gate was designed to be removable so that its size and geometry could be modified with ease. It was felt that this die could be prepared for molding of the various thickness parts by modification of the main center gate (diameter and draft).

The inserts for this mold to provide the various thicknesses consisted of rectangular shims with a 12 in. X 12 in. inside dimension and 15 in. X 15 in. outside dimension. Inserts having thicknesses of 0.125, 0.250, and 0.500 in. were prepared. (It was originally considered that a 0.375-in. part could be made by combining the 0.125 and 0.250-in. inserts. We found, however, that we were not able to obtain adequate sealing between the inserts to utilize such an approach.) These inserts were attached to the rear platen (see Figure 1) by steel dowel pins, to assist in maintaining the internal pressures, with screws to hold them in place. The inserts could then readily be changed simply by removing the screws and prying from the rear platen.

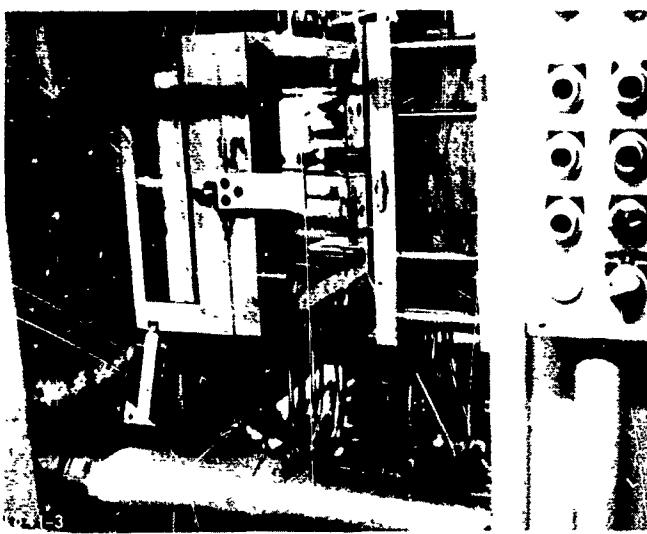


Figure 1. The Die for 12 In. x 12 In. Plates Installed  
In a 20-Ounce Injection Molding Machine.

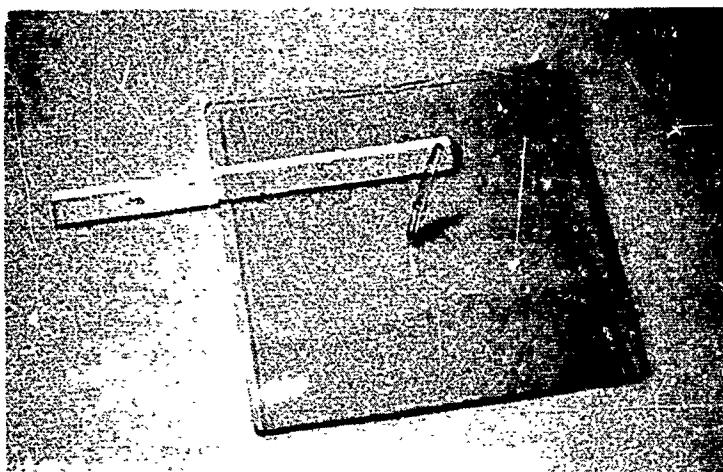


Figure 2. Injection Molded 12 In. x 12 In. x .32 In.  
Polycarbonate Plate (Center Sprue Not Removed).

To provide for the expulsion of air from the die upon molding, vents were machined into the inserts at the sides. These vents consisted of channels approximately 1 in. wide and 0.001 in. deep. The necessity for these vents was one reason that the inserts could not be stacked.

To mold large strain-free parts of polycarbonates, it was necessary that the materials be injected into a mold that was at a temperature above the glass transition temperature of the polycarbonate, i.e., 290°F (145°C). It was necessary that all three plates of the mold have water channels incorporated into them to achieve this type of molding. These water channels were necessary both for steam heating the die and for cooling it to a temperature substantially below its glass transition temperature prior to removal of the part. In molding, the channels of each section of the die were connected to high-pressure steam (130 psi) for heating and switched to city water for cooling.

The injection molding machine for which this die was designed had a 20-ounce rated capacity. A few simple calculations showed that a 12 in. X 12 in. plate, much greater than 1/8 in. in thickness, would contain more than 20 ounces of material. It was, therefore, necessary that the injection-molding machine be of a screw type with an intrusion feature. The intrusion feature provided for an override of the injection screw. Thus, plasticated material could be pumped on demand to the extent desired prior to hydraulically injecting the final charge and application of the compacting pressure.

b. Determination of Molding Conditions for Various Thickness Plates

As was noted in the previous discussion (Section III.3.a), each die had a different capacity and required a different set of molding conditions. In particular, we had to determine the molding conditions for three dies consisting of one basic die with three inserts (1/8, 1/4, and 1/2 in.). The total weight of the 1/8-in. plate was such that the injection molding machine could be used in its normal manner. The 1/4- and 1/2-in. plates weighed greater than 20 ounces; therefore, intrusion was necessary in molding them. In all cases filling of such large parts with their considerable surface area required a hot-die molding technique.

Each plate (thickness) required a different set of molding and mold conditions which consisted of:

- clamp pressure,
- injection pressure,
- mold pressure,
- cylinder temperatures,
- mold temperatures (3-sections),
- cycle time, and
- gate configuration.

We found that it was most important to have the mold heated uniformly throughout and to use the highest temperature available from high-pressure steam. This temperature was normally around 330°F (165°C). Uniformity was obtained only through lengthened die heating times. It was necessary to cool the part to get it out of the die and to cool it as rapidly as possible with available city water. The time that was required to cool the part and the die was affected to a great extent by the lengthened heating cycle. Accordingly, molding of these plates required a substantial amount of time (15 to 30 min), most of which involved the heating and cooling of the die.

While the other portions of the molding cycle required much less time, their values were important. In the case of intrusion, the rate of fill was controlled by the rotation of the screw. While it was important to fill the die as fast as possible, it was required that the die not be filled so fast that air would be entrapped in the parts. The injection pressure provided a final filling of the part which was also adjusted to be completed as fast as possible, while allowing the material to relax into its desired state. The final hold pressure was then applied to pack the part and maintain a significant pressure on it during cooling. The total injection time, including intrusion, was on the order of 5 to 15 seconds.

The clamp pressure, i.e., the pressure required to hold the die together, was adjusted to provide a minimum total pressure commensurate with that required to hold the die together during the application of the high injection and hold pressures. Since it was necessary to allow air to escape from the mold,

and since we did not want to crush any part of the die (a possibility with the 350-ton clamp capacity), clamp pressure was held to a minimum.

To injection-mold a transparent material without loss in optical properties, it was necessary that the material be exposed to the minimum temperatures commensurate with ready extrusion through the system and into the die. Cylinder and nozzle temperatures therefore were adjusted to this minimum to prevent degradation of the polymer. Where it was necessary to provide additional flow (reduced viscosity) to the polymer, the nozzle temperature was increased to give an initial or momentary preheat to the polymer immediately prior to injection. The cylinder temperatures were on the order of 500°F (260°C).

In the handling of any polycarbonate at elevated temperatures, especially in injection molding, it is most important that the polymer be absolutely free from water. All the polycarbonate used in this program, therefore, was predried in a circulating oven at 250°F (121°C) for at least 24 hours prior to use, and the polymer was fed into a dryer hopper. In this hopper, air was continuously circulated at an elevated temperature to assure removal of all water from the polycarbonate. As little as 0.05% of water in the polycarbonate would cause degradation resulting in white spots in the molded part.

Finally, to provide for an adequate rate of fill of the various sizes of parts, the size and geometry of the removable gate was varied. For the most part, it was necessary to increase the size of this gate as the size of the part increased. Also, since the gate, in the form of a cone having a large end of approximately 3/8-in. diameter was required, an initial cut-down section to the gate was required to allow for break away of the part from the screw and nozzle section of the injection molding machine. The problems with this gate increased significantly as the size of the part increased.

A typical injection molding plate with the center sprue removed is shown in Figure 2. Typical molding conditions are shown in Table I, and are valid for this particular die and injection molding machine. These parameters would change if another machine were used, so we shall not discuss the values in detail. If any additional molding is contemplated, the above discussion should be considered in obtaining a desired part.

Table 1

TYPICAL MOLDING CONDITIONS FOR 12 IN. X 12 IN. x 0.5 IN.  
POLYCARBONATE PLATES

Material: Merlon E-271                            Mold: 12 in. x 12 in. plate  
 Lot & Batch: 1000 Nat., J-711-7-019            Inserts: 0.5 in. thick  
 Part No.: 75072                                    Machine: 20 oz. HPM

<u>Cylinder Temperature, °F</u>	<u>Injection, psi and sec.</u>		
Nozzle	525	High	700, 20
Zone 1A	525	Low	700, 300
Zone 1 (front)	525		
Zone 2 (center)	525		
Zone 3 (rear)	525	<u>Cycle Time, sec.</u>	
		Clamp-closed	480
<u>Mold Temperature, °F</u>		Clamp-open	30
		Total	510
Sprue Plate	249		
Center Plate	220		
Rear Plate	220	Worm (Vickers) Setting	
		Injection (travel)	8
		Speed (rotation)	5
<u>Clamp Pressure, psi</u>			
High (closing)	2800	<u>Intrusion time, sec.</u>	
Low (holding)	2800		67

c. Parts Molded

Using the molding conditions discussed in the previous section (III.3.b), thirty-two 0.125-in. thick plates, twenty-four 0.250-in. thick plates, and twelve 0.50-in. thick plates were molded from polycarbonate.

These parts were stored flat in preparation for their use in laminated transparencies. While we attempted to use as little release agent as possible, we found that it was necessary to use a slight amount of calcium stearate for this purpose. This release agent should be removed prior to any attempts at laminating.

4. CHARACTERIZATION OF THE HIGH-CLARITY POLYCARBONATE

The efficacy of the Merlon E-271 polycarbonate in terms of its optical clarity and low-velocity impact resistance was determined following injection molding. Since the low-velocity impact resistance of the polycarbonate can be reduced significantly by exposures at elevated temperatures, a brief examination of annealing conditions was also conducted. The tests were conducted according to standard ASTM and Federal Test Method techniques.

a. Optical Characteristics

A significant portion of the total program was to involve the inclusion of Merlon E-271 polycarbonate for its improved optical clarity over other commercially available polycarbonates. In order to demonstrate this difference, the optical transmittance of the Merlon E-271 and commercially available Merlon® and Lexan® polycarbonate was determined.

The transmittance was measured from 350 to 750 millimicrons wavelength on a Perkin-Elmer Model 350 spectrophotometer. This is a continuously recording device that measures optical transmittance directly.

The optical spectra of 0.125-in. thick pieces of Merlon E-271, Merlon M-50-100 (a natural grade of polycarbonate from Mobay Chemical Company) and Lexan L-141-112 (a natural grade of polycarbonate from General Electric Company) were measured. The results are shown in Figure 3. Comparison of the optical spectra (from 400 to 700 millimicrons) for these materials readily shows the significant improvement in the optical

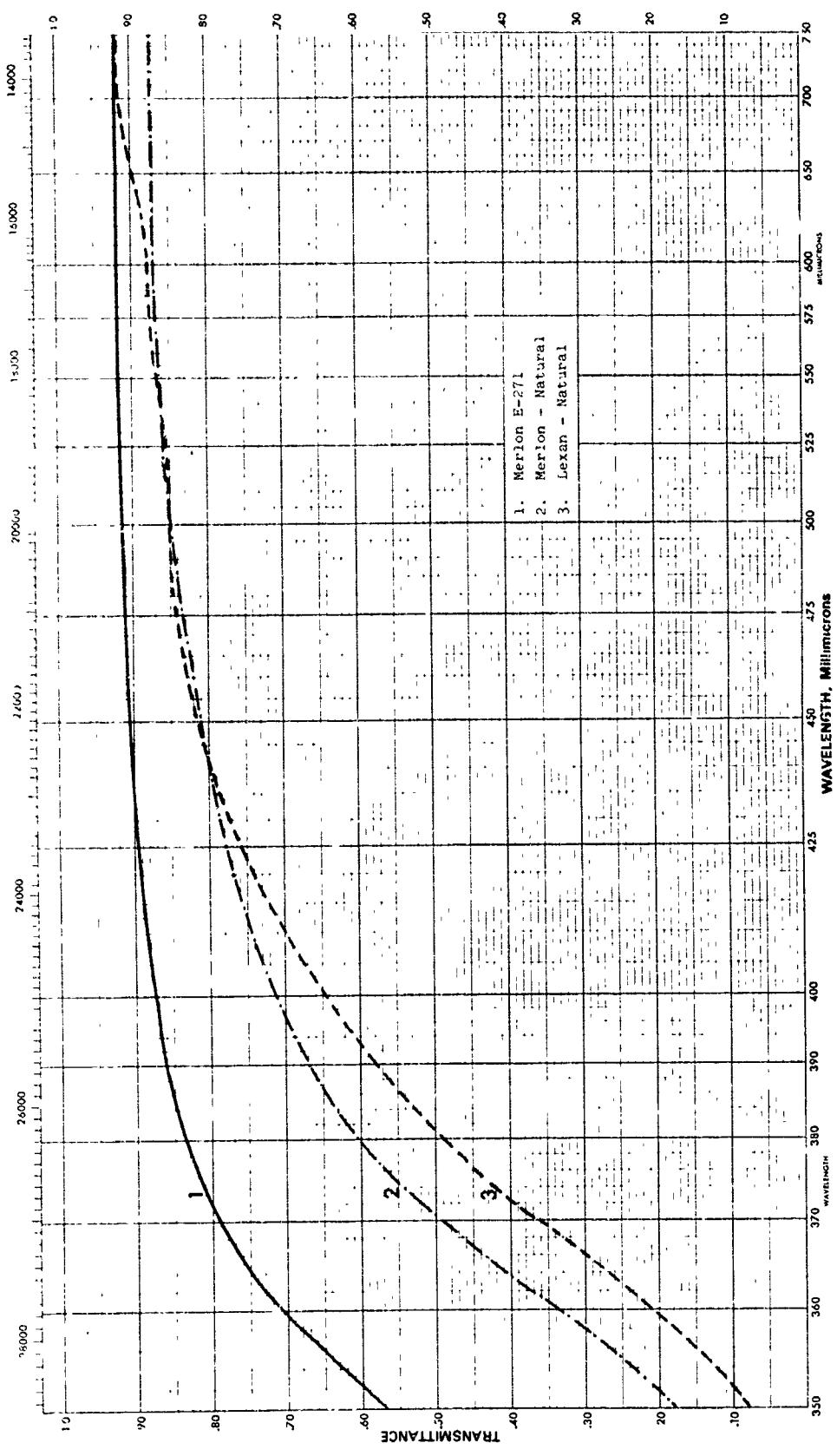


Figure 3. Optical Transmittance of Commercial and High Clarity Polycarbonates.

transmittance of the Merlon E-271. The improvement in optical transmittance was from 5 to 15% on an absolute basis. The E-271 exhibited greater than 88% transmittance over the entire optical range studied.

b. Low Rate (Izod) Impact

Polycarbonate is well-known for its high Izod impact characteristics. Approximately 15 ft-lb/in. of notch is available when tested according to ASTM designation D-256-56. As shown in Figure 4 the Merlon E-271 exhibited this characteristic high-impact resistance.

In laminating glass to polycarbonate, autoclave temperatures of up to 250°F (121°C) and higher are required. Since the high impact characteristics of polycarbonate can be lost through annealing, we determined the temperature at which this annealing would occur. A simple technique (Vicat) for examining the polycarbonate to determine whether it had indeed been exposed to such conditions was also established.

(1) Annealing Temperature Determination

Accordingly, the impact resistance of the Merlon E-271 was determined after specimens had been annealed at various temperatures. Specimens were quenched both in 74°F (23°C) air and in 32°F (0°C) water.

The results of the impact tests, all conducted at 74°F (23°C), following annealing at various temperatures are shown in Figure 4. As shown in Figure 4 the impact strength of this polycarbonate was essentially unaffected by annealing temperatures up to approximately 212°F (100°C). It was also shown that no apparent difference in the impact strength between samples quenched in air or in freezing water existed.

The data shown in Figure 4, however, demonstrated that a drastic reduction in the notched Izod impact strength of the E-271 occurred after the parts had been annealed above 212°F (100°C). The impact strength was reduced to approximately 2 ft-lb/in. of notch, which is in the range of most other non-impact-type plastics. These results also occurred independent of the type of quenching utilized. This effect exists in other polycarbonates, but the temperature at which it occurs depends on the molecular weight of the polymer. The molecular weight of the E-271 polycarbonate is obviously on the low side of commercial polycarbonates.

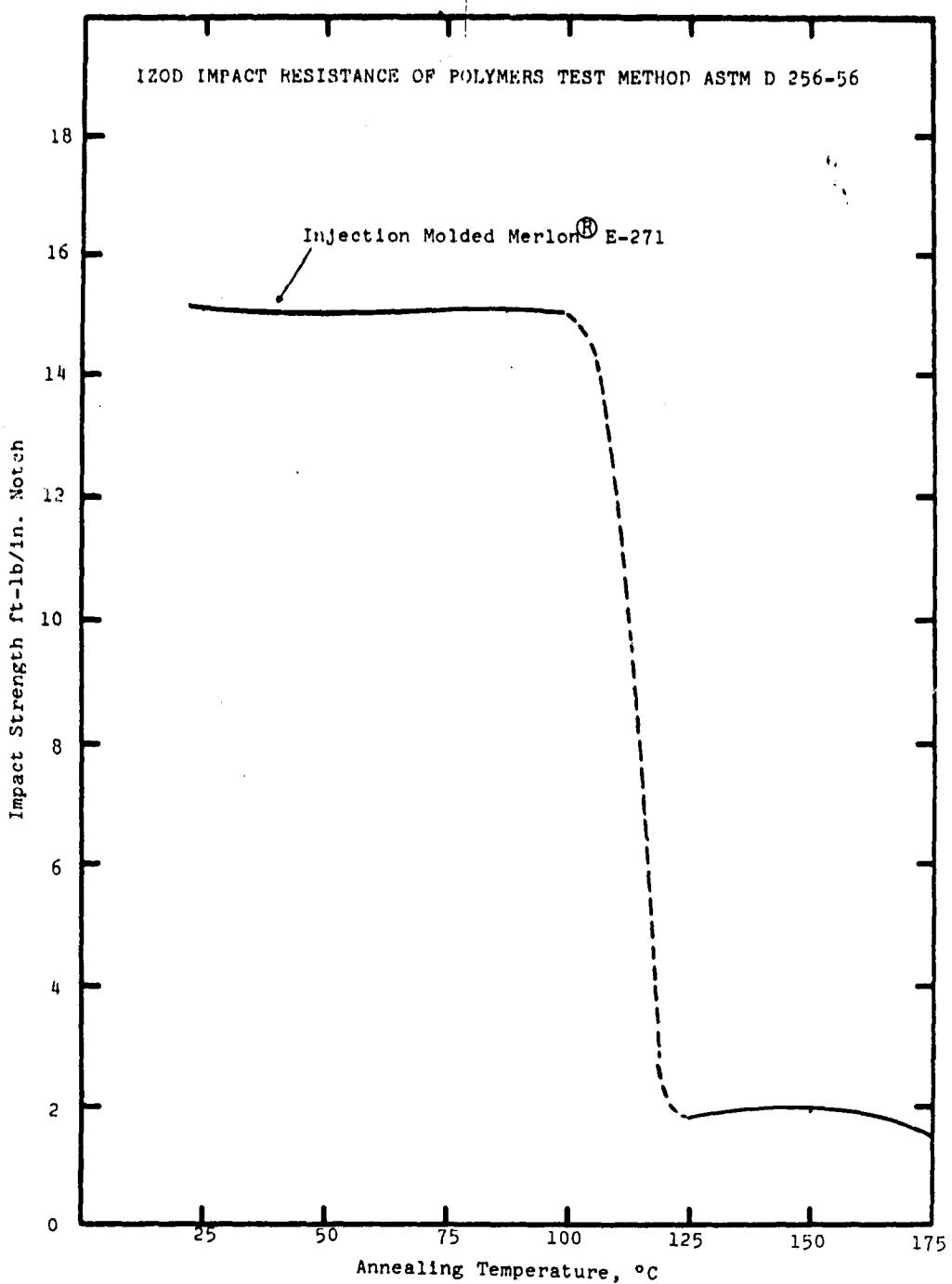


Figure 4. Izod Impact Strength of E-271 Poly-carbonate as a Function of Annealing Temperature.

The loss of impact strength at annealing temperatures above 212°F (100°C) could be attributed to the increase in the crystallinity of the polymer and the absence of a low temperature transition region.

At annealing temperatures of 345°F (175°C) and higher, gas bubbles were formed in the polycarbonate, and the specimens lost their structural integrity. Therefore, handling of the polycarbonate at temperatures much above 300°F (150°C) should be avoided at any time after the polycarbonate plate is formed.

## (2) Identification of Annealing by Vicat Indentation

To provide a technique for determining the degree of annealing of a piece of polycarbonate after molding, laminating, or thermal exposure, we examined the Vicat indentation curves of both annealed and unannealed polycarbonates. The Vicat indentation was performed according to ASTM designation D-1525-58T.

The results of the Vicat indentation for annealed and unannealed polycarbonate samples are shown in Figure 5. The actual Vicat softening point temperature itself was unaffected by the annealing temperature or the method of quenching over the test range from 74°F (23°C) to 345°F (175°C). In all cases the average Vicat softening point temperature was 308°F ±2°F (153°C ±1°C).

However, it was apparent that while the Vicat softening point temperature was independent of annealing, the initial slopes of the indentation versus temperature curves were markedly different. The initial slope of the indentation curve thus was a measure of the degree of annealing in the material. The degree of deviation of the initial slope from a straight line indicated the degree of annealing that had occurred. This annealing correlated to decreased impact resistance.

The Vicat indentation tests, therefore, provided a technique to examine polycarbonate samples, especially after lamination, to determine the nature of the low-rate impact properties without the need of preparing and testing an impact specimen.

## 5. LAMINATING OF TRANSPARENT SHIELDS

Laminating of transparent shields was conducted only to a very limited extent in this program for two reasons. First, considerable time and expense was required to obtain and prepare

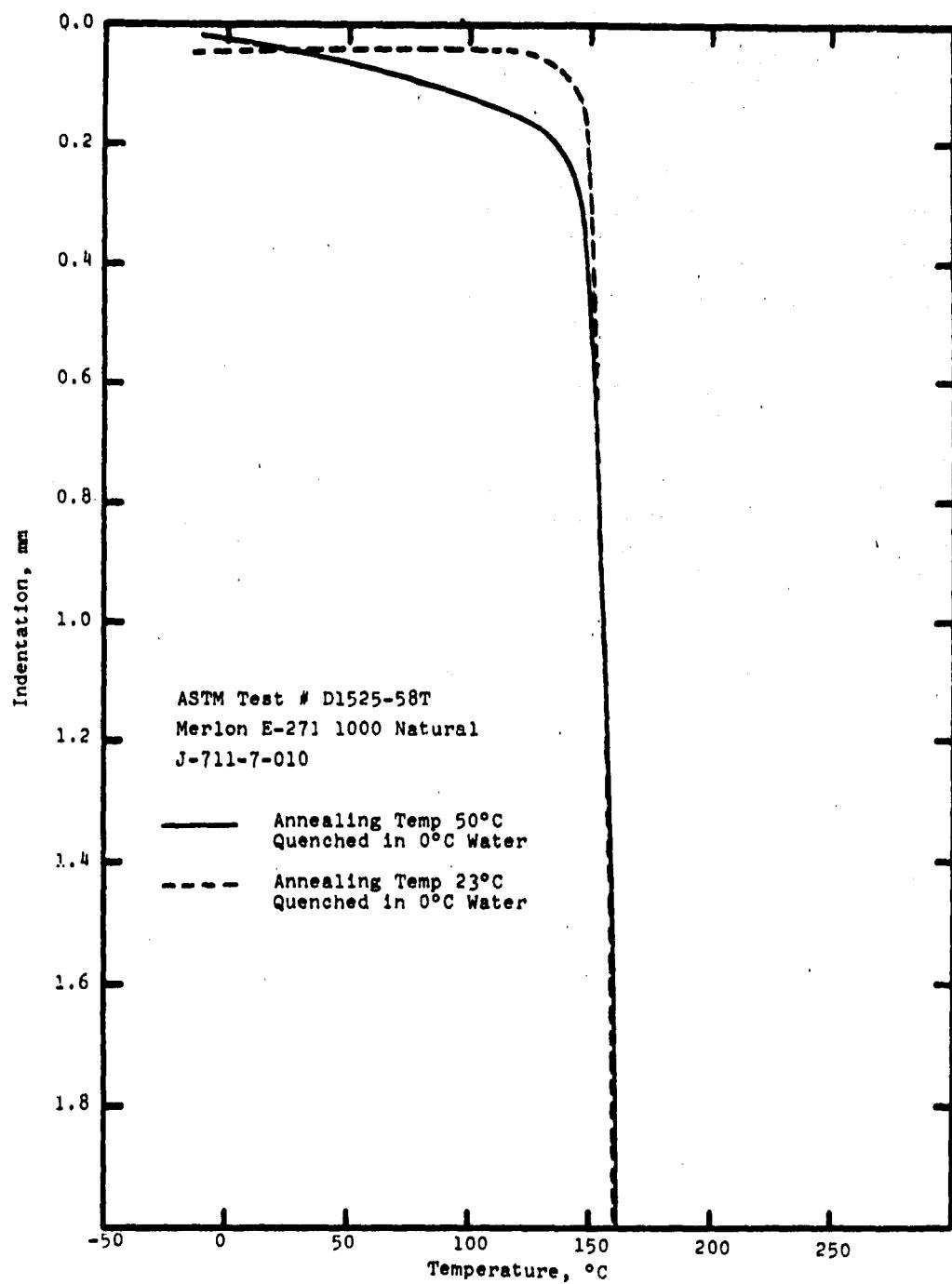


Figure 5. Softening Curve (Vicat) for Merlon E-271 Polycarbonate.

the various materials for the laminate. Second, the proposed adhesives for bonding of the polycarbonate to glass did not perform as well as anticipated.\* The materials obtained for inclusion in transparent laminates, a qualitative description of the laminating conducted, and the optical characterization of some prototype laminates, are described in the remainder of this section.

#### a. Materials

The polycarbonate plates used in this program are described in Sections III.3 and III.4. The main purpose in preparing the high-clarity polycarbonate plates was for use in glass-poly-carbonate laminates. The glass used was Pittsburgh Plate Glass Company's aluminum borosilicate (No. 7428) high-strength glass. This was obtained in 6 in. X 6 in. plates and with thicknesses of 0.25, 0.375, and 0.5 in. We purchased 50, 30, and 20 lites of each thickness, respectively.

The original purpose of this program was to incorporate this high-strength glass into an optimized transparent system. Since an optimized system was not achieved within the duration of the program, this glass was not used except for a few adhesion studies. It has, however, been transferred to the continuing effort for inclusion in the anticipated high-performance transparency.

To conserve this relatively expensive high-strength glass in the laminating and adhesion studies, we used instead 10 in. X 10 in. plates of soda lime glass, obtained by the Air Force from Libbey Owens Ford (100 sheets, 0.25 in.; 60 sheets, 0.375 in.; and 40 sheets, 0.5 in. thick). With the exception of some small specimens made for optical characterization, this was the only glass used during the program. This less expensive glass was to establish laminating conditions, adhesive quality, and optical characteristics without incurring a major material expense.

Adhesives that were investigated included a modified polyvinyl-butylal (Monsanto Company), Texin Thermoplastic Urethane (Mobay Chemical Company), and a thermosetting silicone rubber (General Electric RTV-108).

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\*An excellent ethylene terpolymer adhesive was made available only in the last month of this program and therefore could not be used. The efficacy of this system is a subject of Task 5 under Air Force Contract Number F33615-69-C-1190, AFML-TR-70-144.

The modified polyvinylbutyral (PVB) was a proprietary adhesive developed by Monsanto Company which had been rated as a suitable adhesive for bonding polycarbonate. The system was to gain the effectiveness through substitution of a "non-crazing" plasticizer in the polyvinylbutyral over that used in commercially available PVB. A sample of a laminate of glass and polycarbonate bonded with this modified PVB was sent to us and looked most promising since very little crazing or haze was visible.

The Texin urethane of Mobay Chemical Company is a commercially available thermoplastic sheet that was felt to have suitable adhesive qualities and possibly adequate optical clarity.

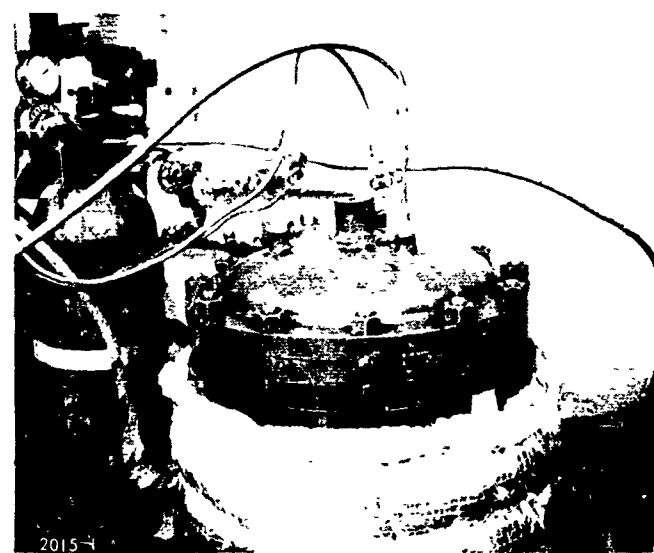
The RTV-108 silicone rubber from General Electric was used purely to combine pieces of glass and polycarbonate for optical clarity measurement. Since silicone had been investigated by others and was not considered to have a good potential, it was not really considered for ultimate use in the system to be developed in this program.

#### b. Techniques

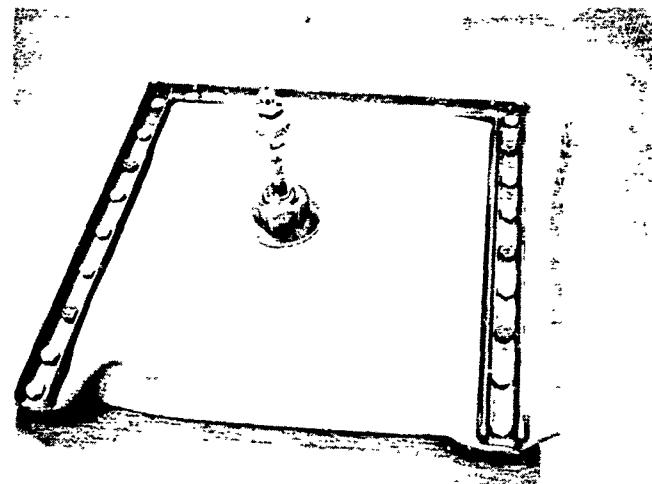
For prototype laminating, where retention of properties was not important, padded hydraulic presses were used to prepare laminates of glass-polycarbonate and glass-glass. This technique, however, was used only to a very limited extent.

In order to achieve uniform laminating and relatively strain-free transparencies (which would exhibit stability with time) we utilized an air-type autoclave. The pressure vessel was jacketed with oil, which was heated by controlled steam. Pressure was provided by high-pressure nitrogen in cylinders, controlled to the desired pressure. This pressure vessel is shown in Figure 6.

It was necessary to provide for removal of entrapped air and moisture from within the laminates. A bagging technique was employed for each specimen in the chamber. One of these bags is shown in Figure 6 with the vacuum line attached. The bagged sample was placed within the air autoclave and evacuated with a vacuum pump. The appropriate temperatures and pressures were then applied. The bag shown is a silicone bag which we found to be necessary for some of the higher temperature laminating. However, we were able to use aluminized Mylar containing a surface film of polyethylene, into which a specimen could be placed, and encapsulated by heat sealing.



(a) High Pressure Autoclave  
and Associated Equipment



(b) Silicone Rubber Vacuum  
Bag for Laminates

Figure 6. Equipment for Pressure-Vacuum Laminating.

The temperature control was maintained by passing regulated steam through the coils in the oil bath for heating and water for cooling. Most importantly, at no time were the specimens exposed to any oil, water, or steam.

c. Laminates

To demonstrate the optical clarity of the polycarbonate to be used in the transparent system, three simple laminates were prepared from 0.125 in. thick pyrex glass plates and 0.125 in. thick injection molded Merlon E-271 polycarbonate. These were bonded with the RTV-108 silicone rubber adhesive. (As noted earlier, the silicone was used purely for convenience in this stage of the program.)

These three laminates consisted of (1) two 0.125 in. thick pyrex glass plates, (2) two 0.125 in. thick pyrex glass plates bonded with RTV-108 silicone rubber adhesive, and (3) two 0.125 in. thick pyrex glass plates bonded by the RTV-108 silicone rubber adhesive to a 0.125 in. thick plate of Merlon E-271 polycarbonate. These three samples were used to demonstrate the optical transmittance allowed by the E-271 polycarbonate.

Figure 7 shows the less than 85% optical transmittance allowed by the glass. This was reduced to approximately 70% transmittance by the silicone adhesive at a wavelength of 400 millimicrons. This reference system was then reduced in transmittance by a maximum of only 6% absolute at the 400 millimicron wavelength due to the polycarbonate.

Various laminates consisting of glass, polycarbonate, and modified PVB were prepared. However, the resultant haze was so obvious that no transmittance measurements were made. To demonstrate the haze that was present in this system, a laminate consisting of two pieces of soda lime glass, two layers of modified PVB, and one layer of polycarbonate was prepared. The laminate was laid-up in the following order: 0.25 in. glass; 30 mils of modified PVB; 0.125 in. polycarbonate; 30 mils of modified PVB; and 0.25 in. glass. The purpose of this particular configuration was to provide at least two modified PVB-polycarbonate interfaces to assist in visual amelioration of the haze problem.

Two views of this laminate are shown in Figure 8 to demonstrate the haze problem. The photo at the top of the page was taken at 0° obliquity to (and through) the specimen onto a back-lighted photographic transparency. The haze and lack of

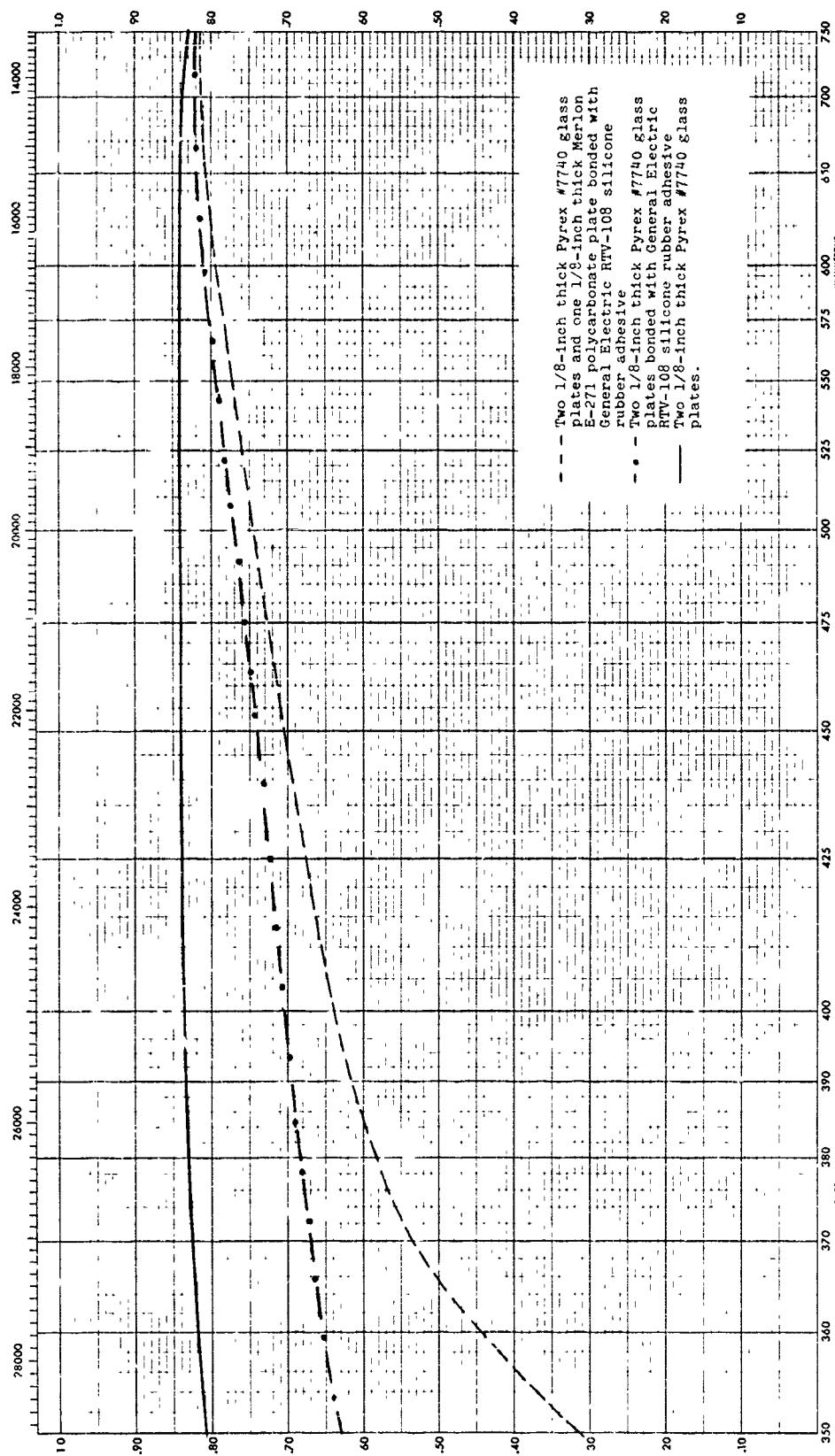
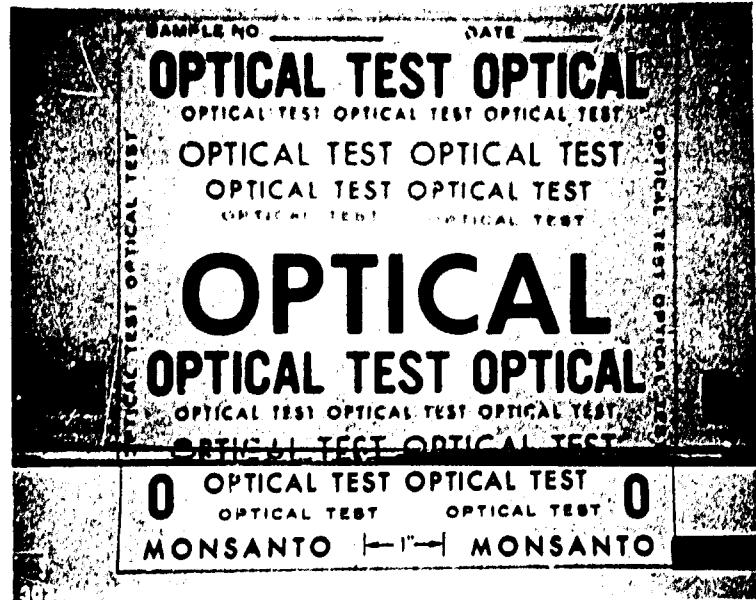


Figure 7. Optical Transmittance of Several Transparent Laminates.



0° Obliquity



45° Obliquity

Figure 8. Glass-Polycarbonate Laminate Bonded With A Modified Polyvinylbutyral Showing the Problem of Haze

definition shown in the mid- and top portions of the picture were due to the craze and haze in the polycarbonate. A difference in the M's at the right and left of the photo was observed. The M on the left was red in color and that on the right blue. The difference in definition was due to this color difference.

The bottom photograph in Figure 8 shows the identical laminate held at a 45° angle to a top-lighted reflecting test pattern. This particular photograph demonstrated the considerable loss of visual acuity due to the haze of the system.

The above comments on the modified polyvinylbutyral adhesive for bonding polycarbonate to glass are negative, but the results do show that a substantial improvement over regular PVB, was made available by this particular adhesive. If commercial PVB had been included in this particular laminate, the image photographed in the bottom of Figure 8 would have been all but obscured, and the definition in the image in the top photo would have been significantly impaired. Importantly, also, the haze evident in Figure 8 did not increase any significant amount over a period of 9 months.

Prototype specimens, in which glass was bonded to polycarbonate and polycarbonate to polycarbonate by Texin thermoplastic urethane, were prepared. Even with extensive drying of the urethane prior to lamination and considerable care in the laminating process, a substantial haze (greater than that shown in Figure 8 for the modified PVB) remained. The adhesion between the urethane and polycarbonate was shown to be poor by simple hammer impact tests, which caused delamination.

While improvements in thermoplastic urethane clarity may become available, and may eventually be of a nature that could be incorporated into a transparent laminate, the art did not allow inclusion during this program. Another significant problem was the fact that laminating temperatures greater than 250°F (120°C) were required to allow flow of the thermoplastic urethane, and this temperature would have had a detrimental effect on the Izod impact characteristics of the polycarbonate. Owing to the lack of ready applicability of the Texin urethane to this system, no more than a minor effort was expended in this program.

## 6. BALLISTIC CHARACTERIZATION AND ANALYSIS

Our ballistic characterization and analysis involved primarily (1) developing the ballistic testing techniques on a range

provided by Monsanto Research Corporation corporate funds, and characterization of the polycarbonate ballistics in terms of thickness and the degree of annealing.

a. Techniques

General views of the ballistic testing facility are shown in Figure 9. This facility was set up based on available space and AMRA MS63-03 specifications.

The propelling weapon was a caliber 0.30 Winchester rifle with a tapered bull barrel. The rifle was rigidly mounted to a dead-weight friction recoil stand loaded so that no bounce occurred. A remote pneumatic firing device was connected to the trigger.

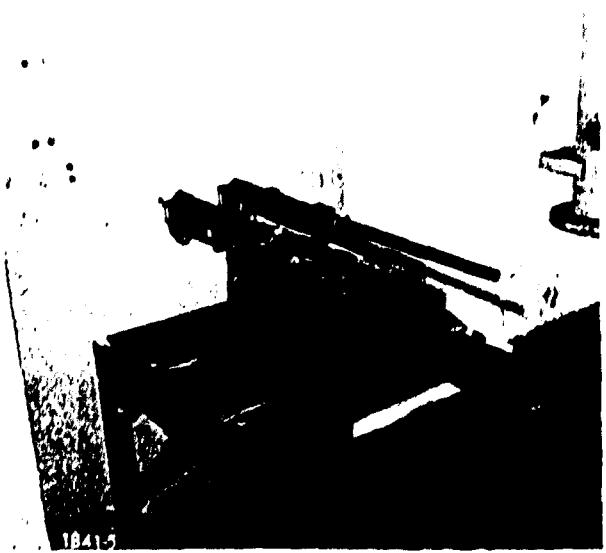
For timing, duplicate resistance screens were spaced 5 feet apart, approximately 5 feet from the barrel of the weapon and 5 feet from the target. The bullet velocity was then determined in duplicate by two Oehler Research Model-10 timers started and stopped by penetration of the screens.

The target holder consisted of an aluminum metal clamp. This was placed within a 0.25 in. steel-walled chamber on a movable stand which was backed up by 2 feet of sand, one inch of plywood, and 1/4 inch of steel plate. The target fragments were confined inside this steel chamber.

The entire ballistic facility was contained within steel-reinforced concrete walls which have been used for the confinement of high-pressure explosions.

Ammunition reloading procedures and techniques were established. This was necessary to provide the various projectile velocities for determining the V50 ballistic limit. The caliber 0.30 rifle was test-fired and the ballistic curve of projectile velocity versus powder charge obtained for both caliber 0.30 ball M2 and AP projectiles.

The V50 ballistic limit, as well as the merit rating, were based on the velocity of the projectiles between the two timer screens and not the striking velocity as specified in report AMRA MS63-03. This difference, however, was not expected to be of any significance, since the distance between the midpoint of the two screens to the target was less than 10 feet. All other test procedures were performed according to the specifications provided in AMRA MS63-03. Precise temperature control was not available to the room so testing was conducted at temperatures of  $74 \pm 6^{\circ}\text{F}$  ( $23^{\circ}\text{C} \pm 3^{\circ}\text{C}$ ).



Caliber .30 and .222 Rifles  
on Friction Mount



Target Box and Timing Screens



Backstop and Target Holder

Figure 9. Ballistic Test Facility.

Where no damage was done to the caliber 0.30 ball M2 or AP type projectiles (in the form of projectile breakup, etc) the only difference between the two projectile impacts was their weight. The AP weighed 165 grains and the ball type 150 grains. Therefore, in testing at equivalent velocities, the V50 ballistic limit for the AP was lower than that for the ball type ammunition.

The kinetic energy necessary to defeat either an AP or a ball projectile should be the same. Thus,

$$KE_{Ball} = KE_{AP}$$

$$KE_{Ball} = \frac{1}{2} M_B V^2 \quad 50B$$

$$KE_{AP} = \frac{1}{2} M_{AP} V^2 \quad 50AP$$

Equating and simplifying,

$$V_{50AP} = [M_B / M_{AP} \times (V_{50B})^2]^{1/2}$$

where  $M_B$ -mass of the ball type projectile,  $M_{AP}$ -mass of the AP type projectile,  $V_{50B}$ -ballistic limit for ball-type projectile,  $V_{50AP}$ -ballistic limit for AP type projectile.

Where hard front surfaces exist, such as in a glass-fronted laminate, and breakup or stripping of a jacket of a projectile occurs, this relationship would not hold. However, testing of this sort was not conducted in this program.

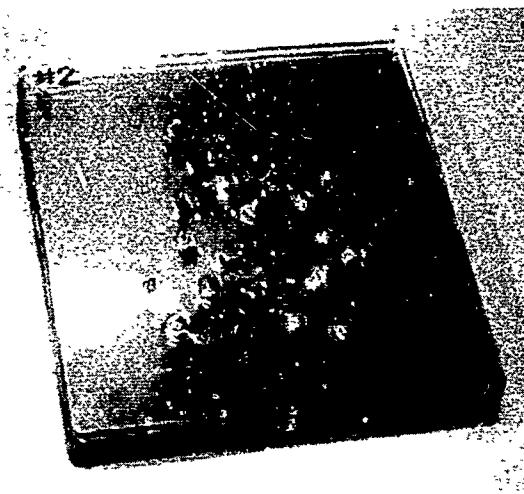
#### b. Ballistic Analysis

To determine the effect of annealing and the loss of low-velocity impact strength on the ballistic performance of polycarbonate, the V50 ballistic limits and merit ratings of various thicknesses of polycarbonate (Merlon M-50 and Lexan 101) were determined. The polycarbonate plates were tested by clamping them in the test fixture as illustrated in Figure 10.

This fixture used for holding these plates for testing was rigidly clamped between an aluminum and plywood plate in the target box at  $0^\circ$  obliquity to the caliber 0.30 rifle. The location of the ballistic impact was readily controlled by adjusting the position of the test fixture within the target box.



(a) Aluminum Test Fixture Containing Unbonded Multiple Ply Polycarbonate Plates.



(b) Ballistically Tested Unbonded Multiple Ply Polycarbonate Plates Demonstrating Multiple Hit Capability.

Figure 10. Polycarbonate Plate Ballistic Test Specimens.

To compare the effect of multiple plates with a homogeneous plate, for a given total thickness, three polycarbonate plate assemblies were prepared and the V50 and merit rating were determined. The first assembly consisted of six polycarbonate plates, each with a nominal thickness of 0.32 in.; the second of 17 plates, each 0.117 in. thick; and the third of 4 plates each, 0.05 in. thick, rigidly clamped together. No adhesive was used between the plates. The results are shown in Table II and illustrated in Figure 11.

Figure 11 shows that the V50 ballistic limit increased as the homogeneity of the polycarbonate assembly increased. That is, a significant improvement in ballistic defeat resulted from a total thickness consisting of the fewest number of plates.

It was demonstrated that the polycarbonate specimens exhibited excellent multi-hit capabilities. This fact is demonstrated in the lower photograph of Figure 10 which shows numerous hits without any fracture of the total specimen. At least 24 projectile impacts were registered in an area of approximately 100 square in. without any catastrophic failure. Damage from the projectiles was confined to small areas immediately around the holes. Photographs of the caliber 0.30 M2 ball and AP projectiles are shown in Figure 12, after having passed through approximately 2 in. of polycarbonate. Note that they are relatively undamaged.

The hole from which the projectiles exited had a diameter smaller than that of the projectile. This was due to the polycarbonate melting around the projectile as it passed through the material, and then solidifying again, partially closing the bullet hole. The greatest damage to the polycarbonate plates occurred when the projectile struck at a velocity very close to the V50 ballistic limit. Under these conditions, some cracking was observed around the exit hole in the rear polycarbonate plate. At slightly higher or lower velocities, however, no spalling at all was observed.

Since annealing of polycarbonate has a catastrophic effect on its low rate (Izod) impact properties (Section III.3.b), it was anticipated that this annealing effect might be extrapolated to high rate impacts (ballistic). An assembly consisting of four 0.5-in. plates of polycarbonate was annealed at 300°F (150°C) for 19 hours. Following this annealing the V50 ballistic limit and merit rating were determined. The results of this test are shown in Table II. Significantly, it was shown that the ballistic performance of the annealed polycarbonate was unchanged from the performance of the unannealed assembly.

Table II  
BALISTIC PERFORMANCE OF UNFACED POLYCARBONATE LAMINATES

Description of Sample	Areal Density, lb/ft	Test <sup>1</sup> Projectile		Assembly Thickness, in.	V <sub>50</sub> <sup>2</sup> B.L., ft/sec	Merit <sup>2, 3</sup> Rating
		0.030	Caliber, Type			
1. 17 plates of Merlon M-50 polycarbonate rigidly clamped together (each plate approx. 0.117 in. thick)	12.62		Ball	2.00	1072	.44
2. 6 plates of Merlon M-50 polycarbonate rigidly clamped together (each plate approx. 0.322 in. thick)	11.50		Ball	1.93	1208	.52
3. 6 plates of Merlon M-50 polycarbonate rigidly clamped together (each plate approx. 0.322 in. thick)	11.50		AP	1.93	1162	.68
4. 17 plates of Merlon M-50 polycarbonate rigidly clamped together, annealed for 16 hours at 130°C	12.62		Ball	2.00	1120	.46
5. 4 plates of Lexan 101 polycarbonate, rigidly clamped together (each plate approx. 0.500 in. thick)	12.50		Ball	2.00	1272	.53
6. Same as item #5 except annealed at 150°C for 19 hours	12.50		Ball	2.00	1240	.52

<sup>1</sup>Projectile Weight - Caliber .30 Ball M2, 150 Grains Propellant, E. I. Dupont IMR 4064 MT16-2208.

<sup>2</sup>Based on the velocity of the projectile midway between the two firing screens. Distance between this point and target - 10 feet zero degree obliquity.

<sup>3</sup>Test Temperatures - Approximately 23°C, 50% relative humidity.

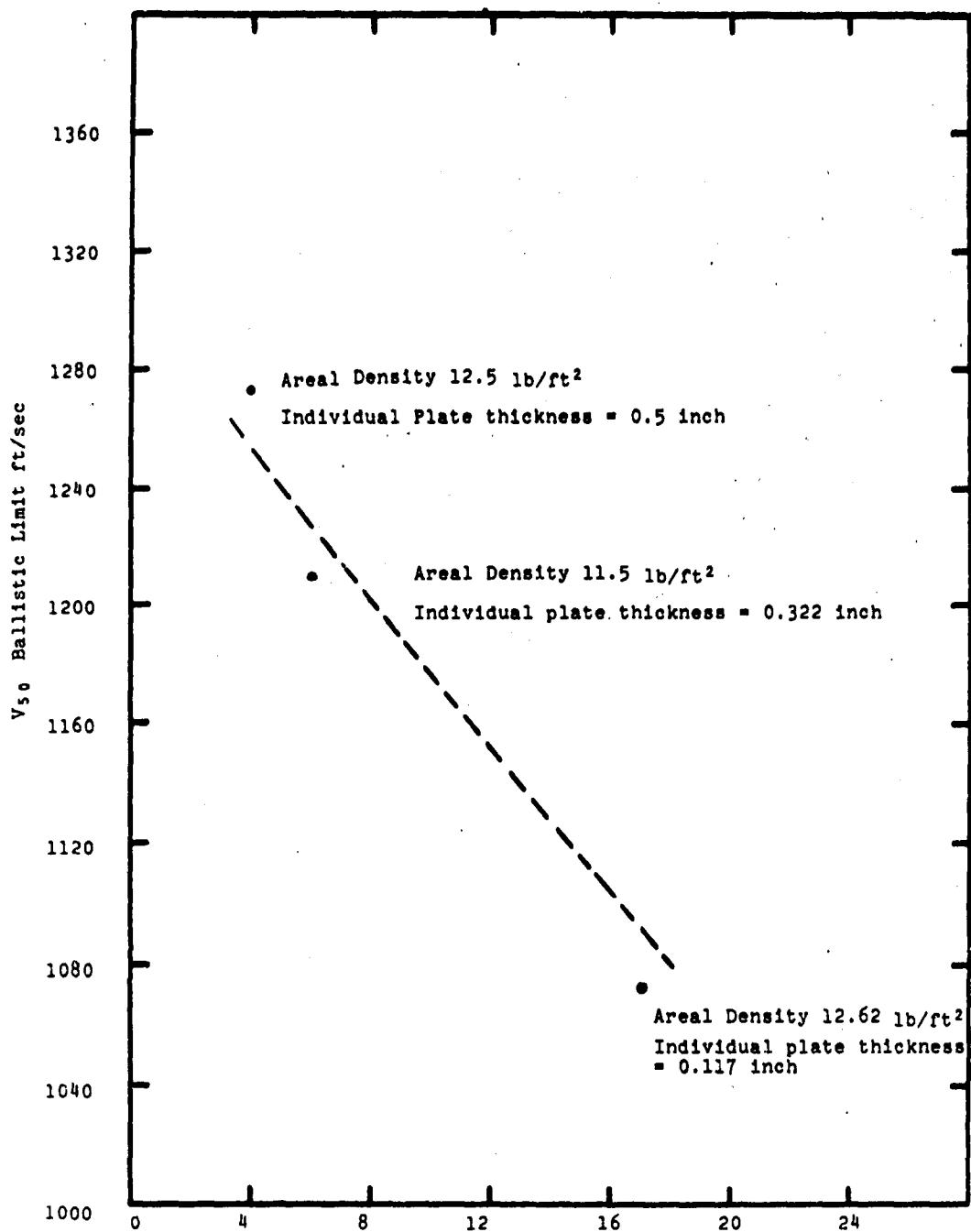


Figure 11. Ballistic Performance of Polycarbonate as a Function of the Number of Unbonded Plies at Equal Total Thickness.

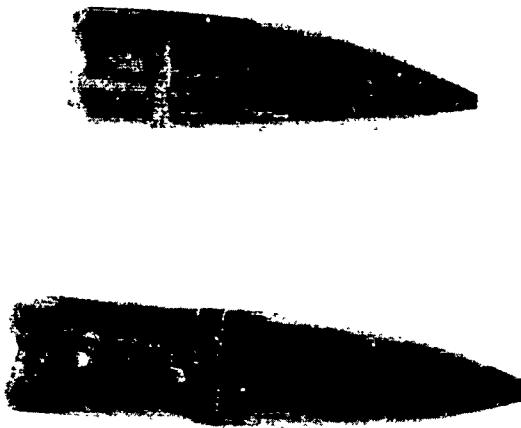


Figure 12. Caliber .30 M2 Ball and AP Projectiles after passing through Approximately 2 In. of Polycarbonate.

Therefore, the characteristics of the polycarbonate which contribute to its excellent Izod impact properties are not significant in its ballistic performance.

At about the same time that we observed this phenomenon, it was reported by the U.S. Army Natick Laboratories that biaxial orientation of polycarbonate provided little significant improvement in ballistic performance, even though standard strength, modulus, and impact characteristics were affected. These, as well as our results, were confirmatory, indicating an unknown mechanism. We hypothesized that since melting occurs upon ballistic impact that annealing or orientation is invalidated.

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11 SUPPLEMENTARY NOTES	12 SPONSORING MILITARY ACTIVITY Air Force Materials Laboratory Wright-Patterson Air Force Base 45433	
13 ABSTRACT  A program was conducted to make available to the Air Force the most advanced materials for use in transparent armor. The materials of interest were a high strength aluminum borosilicate glass, a high clarity polycarbonate, and transparent adhesives for bonding the glass to the polycarbonate.  The aluminum borosilicate glass was made available in various thicknesses up to 1/2 inch by PPG Industries. The polycarbonate was an injection moldable grade from Mobay Chemical Company, described as Meron® E-271. It was necessary to determine injection molding cycles and to injection mold 12 inch by 12 inch plates in thicknesses from 1/8 inch up to 1/2 inch.  Two transparent adhesives were examined. These consisted of a modified polyvinylbutyral (PVB) from Monsanto Company and a thermoplastic urethane from Mobay Chemical Company. Unfortunately, neither adhesive was found to be suitable for the application since haze was evident in laminates and could not be removed. Efforts to refine either one of these materials were discontinued since a more promising material became available at the end of the program.  A minor amount of ballistic testing was conducted on the polycarbonate and significantly it was shown that ballistic performance was not related to low rate impact properties. The implication of these results is that orientation of a thermoplastic is of little benefit and annealing is not detrimental to ballistic performance. It was also shown that a single homogeneous plate had better ballistic performance than multiple plies of polycarbonate.		

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